

09/892177

Patent

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IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Patentee(s): Saleh A. Elomari, *et al.*

Patent No. 6,720,468 B2 (S/N 09/892,177)

Issued April 13, 2004

For: PROCESS FOR THE REMOVAL OF CONJUGATED  
OLEFINS FROM A MONOOLEFIN STREAM

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Group Art Unit: 1764

Examiner: Tam M. Nguyen

Confirmation No.: 1734

**Certificate**

**JAN 27 2005**

**of Correction**

Certificate of Correction Branch  
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**REQUEST FOR CERTIFICATE OF CORRECTION**

Commissioner:

Patentees hereby request correction of the mistake as set out in the attached draft Certificate of Correction and that a Certificate of Correction be issued pursuant to 37 C.F.R. §1.322.

Patentees submitted an Amendment and Response on December 20, 2002 which was entered by the Examiner. However, when submitting a subsequent Amendment and Response to the Final Office Action of March 7, 2003, the word "removing" was inadvertently left out from claim 15 (now renumbered as claim 13) while accepting the previous amendments to said claim. Copies of both documents are enclosed for reference.

A check in the amount of \$100.00 is enclosed in payment of the appropriate fee associated with this communication.

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Date: 1-18-05

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Respectfully submitted,

*Rodney B. Carroll*  
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# UNITED STATES PATENT AND TRADEMARK OFFICE

## CERTIFICATE OF CORRECTION

**PATENT NO. : 6,720,468 B2**

**DATED : April 13, 2004**

**INVENTORS : Elomari et al.**

It is certified that error appears in the above-identified patent and that said Letters Patent is hereby corrected as shown below:

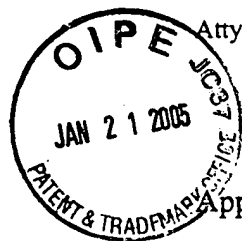
In Column 9, claim 13, line 36, insert "removing" between "said" and "is"

MAILING ADDRESS OF SENDER:

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5700 Granite Parkway, #330  
Plano, Texas 75024

PATENT NO. 6, 720,468 B2

JAN 28 2005



Atty Docket: 60/215583US1 (4081-01200)...

## IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicants: Saleh A. Eloman

Serial No.: 09/892,177

Filed: June 26, 2001

For: PROCESS FOR THE REMOVAL OF  
CONJUGATED OLEFINS FROM A  
MONOOLEFIN STREAM

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Group Art Unit: 1764

Examiner: Tam M. Nguyen

Confirmation No. 1734

Box NON-FEE AMENDMENT  
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### AMENDMENT AND RESPONSE

Commissioner:

In response to the Office Action dated September 20, 2002, please amend the above-identified application as follows. The changes made are shown by underlining the added text and striking through the deleted text. A clean version reflecting the amendments is provided in Exhibit A. A complete set of the currently pending claims is provided as Exhibit B.

#### **In the Claims:**

Please amend the claims as follows:

1. (Amended) A process for ~~the separation of conjugated olefins and monoolefins,~~  
purifying a monoolefin stream, comprising:

contacting a ~~fluid~~ monoolefin stream comprising one or more monoolefins and ~~conjugated olefins~~ with a Diels-Alder dienophile to ~~provide a fluid comprising a~~ Diels-Alder adduct and monoolefins; convert one or more conjugated olefins present in the monoolefin stream to a Diels-Alder adduct;

and removing the Diels-Alder adduct from the monoolefin stream, thereby purifying the monoolefin stream such that it comprises ~~recovering a resulting monoolefin-~~ ~~containing fluid comprising~~ less than about 50 parts per million (ppm) conjugated olefins.

2. Cancelled.
5. (Amended) A process according to claim 1 wherein said conjugated olefins comprise at least about ~~about~~ 4 carbon atoms per molecule and no more than about 10 carbon atoms per molecule.
12. (Amended) A process according to claim 1 wherein said ~~resulting monoolefin-~~ ~~containing fluid~~ purified monoolefin stream comprises less than about 25 parts per million conjugated olefins.
13. (Amended) A process according to claim 1 wherein said ~~resulting monoolefin-~~ ~~containing fluid~~ purified monoolefin stream comprises less than about 10 parts per million conjugated olefins.
14. Cancelled.
15. (Amended) A process according to claim ~~14~~ 1 wherein said ~~separating means~~ removing is selected from the group consisting of distillation, adsorption, membrane separation, and combinations thereof.

16. (Amended) A process according to claim 1 wherein said ~~recovering~~ removing is conducted using reactive distillation.

Please add new claims 19, 20, 21, and 22 as follows.

19. The process according to claim 1 wherein said Diels-Alder dienophile is generally represented by the formula:

$R^1R^2C = CR^3R^4$  where

$R^1 = H, C(=O)OR^5, C(=O)R^6, C(=O)NR^7R^8, CN, C_1 \text{ to } C_{30} \text{ alkyl, and aromatic,}$

$R^2 = H, C(=O)OR^5, C(=O)R^6, C(=O)NR^7R^8, CN, C_1 \text{ to } C_{30} \text{ alkyl, and aromatic,}$

$R^3 = H, C(=O)OR^5, C(=O)R^6, C(=O)NR^7R^8, CN, C_1 \text{ to } C_{30} \text{ alkyl, and aromatic,}$

$R^4 = H, C(=O)OR^5, C(=O)R^6, C(=O)NR^7R^8, CN, C_1 \text{ to } C_{30} \text{ alkyl, and aromatic,}$

$R^5 = C_1 \text{ to } C_{10} \text{ alkyl, aromatic, and } (H)C=CH_2,$

$R^6 = C_1 \text{ to } C_{10} \text{ alkyl, aromatic, and } (H)C=CH_2,$

$R^7 = C_1 \text{ to } C_{10} \text{ alkyl, aromatic, and}$

$R^8 = C_1 \text{ to } C_{10} \text{ alkyl, and aromatic.}$

20. The process according to claim 1 wherein said Diels-Alder dienophile is generally represented by the formula:

$R^1C \equiv CR^2$  where

$R^1 = H, C(=O)OR^3, C(=O)R^4, C(=O)NR^5R^6, CN, C_1 \text{ to } C_{10} \text{ alkyl, and aromatic,}$

$R^2 = H, C(=O)OR^3, C(=O)R^4, C(=O)NR^5R^6, CN, C_1 \text{ to } C_{10} \text{ alkyl, and aromatic}$

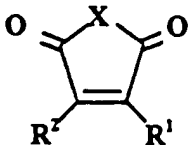
$R^3 = C_1 \text{ to } C_{10} \text{ alkyl, and aromatic,}$

$R^4 = H, C_1 \text{ to } C_{10} \text{ alkyl, and aromatic,}$

$R^5 = C_1 \text{ to } C_{10} \text{ alkyl, and aromatic, and}$

$R^6 = C_1 \text{ to } C_{10} \text{ alkyl, and aromatic.}$

21. The process according to claim 1 wherein said Diels-Alder dienophile is generally represented by the formula:

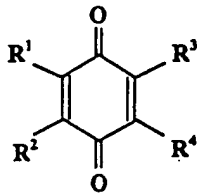


where X = O, N, and S,

R<sup>1</sup> = H, C<sub>1</sub> to C<sub>10</sub> alkyl, and aromatic, and

R<sup>2</sup> = H, C<sub>1</sub> to C<sub>10</sub> alkyl, and aromatic.

22. The process according to claim 1 wherein said Diels-Alder dienophile is generally represented by the formula:



where

R<sup>1</sup> = H, C<sub>1</sub> to C<sub>10</sub> alkyl, and aromatic, and (H)C=CH<sub>2</sub>,

R<sup>2</sup> = H, C<sub>1</sub> to C<sub>10</sub> alkyl, aromatic, and (H)C=CH<sub>2</sub>,

R<sup>3</sup> = H, C<sub>1</sub> to C<sub>10</sub> alkyl, aromatic, and (H)C=CH<sub>2</sub>, and

R<sup>4</sup> = H, C<sub>1</sub> to C<sub>10</sub> alkyl, aromatic, and (H)C=CH<sub>2</sub>.

### **Remarks**

Claims 1, 5, 12, 13, 15, and 16 have been amended; claims 2 and 14 have been canceled; and claims 19-22 have been added. Therefore, claims 1, 3-13, and 15-22 are currently pending. Applicants hereby request further examination and reconsideration of the presently claimed application.

### **Claims Rejection – 35 U.S.C. § 112**

Claims 1-18 have been rejected under 35 U.S.C. § 112, second paragraph, as being indefinite. Claim 2 has been canceled, and new claims 19-22 have been added that include the general formulas for ethylenes, acetylenes, and cyclics as suggested by the Office Action.

Applicants respectfully disagree with the Examiner's comment that the expressions "derivatives of maleic anhydride" and "derivatives of benzoquinone" render claim 3 indefinite. The term "derivative" is well known in the art as meaning a compound obtained from another compound by a chemical process. Therefore, a person of ordinary skill in the art would understand that the expression "derivatives of maleic anhydride" refers to compounds formed by subjecting maleic anhydride to known chemical processes. A person of ordinary skill in the art would also understand that the expression "derivatives of benzoquinone" refers to compounds formed by subjecting benzoquinone to known chemical processes. Applicants therefore submit that these expressions particularly point out and distinctly claim the subject matter that the Applicants regard as the invention.

The Office Action fails to provide any reasons for the Section 112 rejection of claim 1 and claims 4-18. In view of the foregoing, Applicants request that the 35 U.S.C. § 112, second paragraph, rejection of claims 1-18 be withdrawn.

### Claims Rejection – 35 U.S.C. § 103

Claims 1-18 have been rejected under 35 U.S.C. § 103(a) as being unpatentable over U.S. Patent No. 5,300,126 to Brown et al. (hereinafter “Brown”). In response to the obviousness rejection, Applicants respectfully submit that Brown does not establish a *prima facie* case of obviousness as to the currently pending claims.

To establish a *prima facie* case of obviousness, the prior art reference (or references when combined) must teach or suggest all the claim limitations (MPEP 2142). Claim 1 in its amended form provides a process for purifying a monoolefin stream in which one or more conjugated olefins present in the monoolefin stream are converted to a Diels-Alder adduct, followed by removing the Diels-Alder adduct from the monoolefin stream, thereby purifying the monoolefin stream. Brown does not teach or suggest purifying a monoolefin stream by the two-step process of conversion of one or more conjugated olefins to a Diels-Alder adduct followed by the removal of the Diels-Alder adduct. In contrast, Brown discloses converting conjugated dienes (which deactivate acidic catalysts) in an olefinic hydrocarbon feedstream to a Diels-Alder adduct (which do not deactivate acidic catalysts). See Brown, abstract. Brown neither teaches nor suggests removing a Diels-Alder adduct from a monoolefin stream. In contrast, Brown discloses that the olefinic hydrocarbon feedstream containing the adduct is subjected to catalytic conversion. See Brown, column 3, lines 18-22. Given that the resulting adduct does not deactivate the catalyst, Brown does not provide any motivation for removing the adduct from the feedstream. Accordingly, Applicants respectfully submit that independent claim 1 is not obvious in view of the prior art. Likewise, claims 3-13 and 15-22, which depend from claim 1, are not obvious in view of the prior art. In view of the



foregoing, Applicants request that the 35 U.S.C. § 103(a), second paragraph, rejection of claims 1-18 be withdrawn.

The Commissioner is hereby authorized to charge payment of any further fees associated with any of the foregoing papers submitted herewith, or to credit any overpayment thereof, to Deposit Account No. 50-1515 , Conley Rose, P.C.

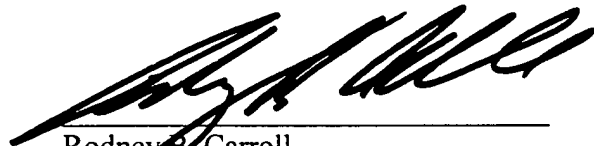
Applicants respectfully submit that the present application is now in condition for allowance. If the Examiner has any questions or comments or otherwise feels it would be helpful in expediting the application, he is encouraged to telephone the undersigned at (972) 731-2288.

Respectfully submitted,

CONLEY ROSE, P.C.

Date: December 20, 2002

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ATTORNEY FOR APPLICANTS

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicants: Saleh A. Elomari, et al.

Serial No.: 09/892,177

Filed: June 26, 2001

For: PROCESS FOR THE REMOVAL OF  
CONJUGATED OLEFINS FROM A  
MONOOLEFIN STREAM

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Group Art Unit: 1764

Examiner: Tam M. Nguyen

Confirmation No. 1734

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*Edith Shek*

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**AMENDMENT AND RESPONSE  
TO FINAL OFFICE ACTION OF MARCH 7, 2003**

Sir:

In response to the Final Office Action dated March 7, 2003, Applicants respectfully request the amendments to the above-identified application as follows. The changes made are shown by underlining the added text and striking through the deleted text.

**Amendments to the Claims begin on page 2 of this paper.**

**Arguments/Remarks begin on page 7 of this paper.**

## AMENDMENTS TO THE CLAIMS

### *Listing of Claims:*

1. (Currently Amended) A process for purifying a monoolefin stream, comprising:  
contacting a gaseous monoolefin stream comprising one or more monoolefins with a Diels-Alder dienophile to convert one or more conjugated olefins present in the monoolefin stream to a Diels-Alder adduct;  
and removing the Diels-Alder adduct from the monoolefin stream, thereby purifying the monoolefin stream such that it comprises less than about 50 parts per million (ppm) conjugated olefins.
2. Canceled.
3. (Currently Amended) A process according to claim ~~2~~1 wherein said Diels-Alder dieneophile is selected from the group consisting of maleic anhydride, ~~derivatives of maleic anhydride, benzoquinone, derivatives of benzoquinone, dialkyl fumarates, dialkyl maleates, dialkylacetylenedicarboxylates, and combinations thereof.~~
4. (Original) A process according to claim 3 wherein said Diels-Alder dieneophile is maleic anhydride.
5. (Previously Presented) A process according to claim 1 wherein said conjugated olefins comprise at least about 4 carbon atoms per molecule and no more than about 10 carbon atoms per molecule.
6. (Original) A process according to claim 5 wherein said conjugated olefins comprise at least about 4 carbon atoms per molecule and no more than about 8 carbon atoms per molecule.

7. (Original) A process according to claim 1 wherein said conjugated olefins are selected from the group consisting of 1,3-butadiene, 1,3-pentadiene, 1,3-hexadiene, 2,4-hexadiene, 1,3,5-hexatriene, 1,3-heptadiene, 2,4-heptadiene, 1,3,5-heptatriene, 1,3-octadiene, 2,4-octadiene, 3,5-octadiene, 1,3,5-octatriene, 2,4,6-octatriene, 1,3,5,7-octatetriene, 1,3-nonadiene, 2,4-nonadiene, 3,5-nonadiene, 1,3,5-nonatriene, 2,4,6-nonatriene, 1,3,5,7,nonatetraene, 1,3-decadiene, 2,4-decadiene, 3,5-decadiene, 4,6-decadiene, 1,3,5-decatriene, 2,4,6-decatriene, 3,5,7-decatriene, 1,3,5,7-decatetraene, 2,4,6,8-decatetraene, 1,3,5,7,9-decapentaene, and combinations thereof.
8. (Original) A process according to claim 7 wherein said conjugated olefins are selected from the group consisting of 1,3-butadiene, 1,3-pentadiene, 1,3-hexadiene, 1,3-heptadiene, 1,3-octadiene, 1,3-nonadiene and 1,3-decadiene.
9. (Original) A process according to claim 1 wherein said monoolefins comprise normal alpha olefins.
10. (Original) A process according to claim 1 wherein said monoolefins are selected from the group consisting of 1-butene, 1-pentane, 1-hexene, 1-heptene, 1-octene, 1-nonene, 1-decene, and combinations thereof.
11. (Original) A process according to claim 10 wherein said monoolefins are selected from the group consisting of 1-butene, 1-pentene, 1-hexene, and combinations thereof.
12. (Previously Presented) A process according to claim 1 wherein said purified monoolefin stream comprises less than about 25 parts per million conjugated olefins.
13. (Previously Presented) A process according to claim 1 wherein said purified monoolefin stream comprises less than about 10 parts per million conjugated olefins.

14. Canceled.
15. (Previously Presented) A process according to claim 1 wherein said is selected from the group consisting of distillation, adsorption, membrane separation, and combinations thereof.
16. (Previously Presented) A process according to claim 1 wherein said removing is conducted using reactive distillation.
17. (Original) A process according to claim 1 wherein said monoolefins are 1-butene and said conjugated olefins are 1,3-butadiene.
18. (Original) A process according to claim 17 wherein said dienophile is maleic anhydride.
19. (Previously Presented) The process according to claim 1 wherein said Diels-Alder dienophile is generally represented by the formula:  

$$R^1R^2C = CR^3R^4$$
where  
 $R^1 = H, C(=O)OR^5, C(=O)R^6, C(=O)NR^7R^8, CN, C_1 \text{ to } C_{30} \text{ alkyl, and aromatic,}$   
 $R^2 = H, C(=O)OR^5, C(=O)R^6, C(=O)NR^7R^8, CN, C_1 \text{ to } C_{30} \text{ alkyl, and aromatic,}$   
 $R^3 = H, C(=O)OR^5, C(=O)R^6, C(=O)NR^7R^8, CN, C_1 \text{ to } C_{30} \text{ alkyl, and aromatic,}$   
 $R^4 = H, C(=O)OR^5, C(=O)R^6, C(=O)NR^7R^8, CN, C_1 \text{ to } C_{30} \text{ alkyl, and aromatic,}$   
 $R^5 = C_1 \text{ to } C_{10} \text{ alkyl, aromatic, and } (H)C=CH_2,$   
 $R^6 = C_1 \text{ to } C_{10} \text{ alkyl, aromatic, and } (H)C=CH_2,$   
 $R^7 = C_1 \text{ to } C_{10} \text{ alkyl, aromatic, and}$   
 $R^8 = C_1 \text{ to } C_{10} \text{ alkyl, and aromatic.}$
20. (Previously Presented) The process according to claim 1 wherein said Diels-Alder dienophile is generally represented by the formula:

$R^1C \equiv CR^2$  where

$R^1 = H, C(=O)OR^3, C(=O)R^4, C(=O)NR^5R^6, CN, C_1 \text{ to } C_{10} \text{ alkyl, and aromatic,}$

$R^2 = H, C(=O)OR^3, C(=O)R^4, C(=O)NR^5R^6, CN, C_1 \text{ to } C_{10} \text{ alkyl, and aromatic}$

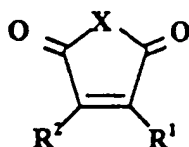
$R^3 = C_1 \text{ to } C_{10} \text{ alkyl, and aromatic,}$

$R^4 = H, C_1 \text{ to } C_{10} \text{ alkyl, and aromatic,}$

$R^5 = C_1 \text{ to } C_{10} \text{ alkyl, and aromatic, and}$

$R^6 = C_1 \text{ to } C_{10} \text{ alkyl, and aromatic.}$

21. (Previously Presented) The process according to claim 1 wherein said Diels-Alder dienophile is generally represented by the formula:

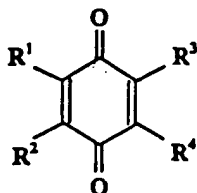


where  $X = O, N, \text{ and } S,$

$R^1 = H, C_1 \text{ to } C_{10} \text{ alkyl, and aromatic, and}$

$R^2 = H, C_1 \text{ to } C_{10} \text{ alkyl, and aromatic.}$

22. (Previously Presented) The process according to claim 1 wherein said Diels-Alder dienophile is generally represented by the formula:



where

$R^1 = \text{H, C}_1 \text{ to C}_{10} \text{ alkyl, and aromatic, and (H)C=CH}_2,$

$R^2 = \text{H, C}_1 \text{ to C}_{10} \text{ alkyl, aromatic, and (H)C=CH}_2,$

$R^3 = \text{H, C}_1 \text{ to C}_{10} \text{ alkyl, aromatic, and (H)C=CH}_2, \text{ and}$

$R^4 = \text{H, C}_1 \text{ to C}_{10} \text{ alkyl, aromatic, and (H)C=CH}_2.$

## ARGUMENTS/REMARKS

Claims 1, 3-13, and 15-22 are currently pending, with claims 1 and 3 having been amended above.

### Claims Rejection – 35 U.S.C. § 112

Claim 3 has been amended to remove the objected to terms.

### Claims Rejection – 35 U.S.C. § 103

Claims 1, 3-13 and 15-22 stand rejected under 35 USC 103(a) as obvious over *Brown* (5,300,126). Applicants and the Examiner agree on the fact that *Brown* does not specifically disclose the step of separating Diels-Alder adduct from the product mixture. Applicants respectfully submit that the Examiner has failed to establish a prima facie case of obviousness as there is no teaching or suggestion to modify *Brown* to remove the Diels-Alder adduct, and in fact there is express teaching against such.

The *Brown* reference is directed to preparation of gasoline blending components having desirable oxygen and octane values, for example ethers such as MTBE. In preparing such gasoline blending components, a liquid olefinic hydrocarbon feedstream such as FCC gasoline comprising C<sub>5</sub>-C<sub>6</sub> is subjected to catalytic conversion via contact with an acidic catalyst. Conjugated dienes present in the olefinic hydrocarbon feedstock are a poison to the acidic catalyst. *Brown* teaches conversion of the conjugated dienes to the corresponding Diels-Alder adduct, which are not poison to the acidic catalyst. Given that the resulting adduct does not deactivate the catalyst, *Brown* does not provide any motivation for removing the adduct from the product mixture. *Brown* is directed to the treatment of liquid feedstreams that are suitable for gasoline blending components, whereas amended claim 1 recites purification of a gaseous mono olefin stream (as shown in the Examples), which is not



suitable for gasoline blending. Furthermore, *Brown* repeatedly and specifically teaches against removing the adduct because the adduct is beneficial for gasoline blending purposes:

...reacting the dienes with one or more dienophiles to form the corresponding Diels-Alder adduct, followed by catalytic conversion of the olefinic hydrocarbon feedstock containing the adduct. See Abstract. See also col. 3, lines 14-24.

...it has been discovered that the adduct, particularly those adducts formed with MA [maleic anhydride], is in the gasoline boiling range and contributes usefully to the oxygen enrichment of the gasoline and to octane value. See Abstract. See also col. 3, lines 31-37.

The invention further encompasses a novel composition of high octane value gasoline having an enhanced oxygen content. The novel composition consists of C5+ gasoline boiling range hydrocarbons containing alkyl substituted tetrahydrophthalic anhydride [i.e., the adduct] or C1-C4 esters thereof. See col. 3, line 67 – col. 4, line 4.

One of the significant features of the process of the invention is the fact that the treated feedstock containing the

diene/dienophile adduct can be subjected to etherification of isoolefins in the feedstock without separation of the adduct.

See col. 9, lines 3-7.

It is a matter of considerable surprise and novelty in the present invention that the tetrahydrophthalic anhydride adducts prepared as described above in a gasoline boiling range hydrocarbon mixture result in a unique composition that displays both high octane value as well as supplementing the oxygen content of the gasoline. See col. 9, lines 23-28.

Given that usefulness of the adduct for gasoline blending is a “significant feature” and of “considerable surprise and novelty”, *Brown* unequivocally teaches against the removal of the adduct. Thus, one skilled in the art would not be motivated to remove the adduct because *Brown* teaches away from such, and therefore Applicants respectfully submit that *Brown* does not provide a prima facie case of obviousness against the pending claims.

## CONCLUSION

Applicants respectfully submit that the present application as amended is now in condition for allowance. If the Examiner has any questions or comments or otherwise feels it would be helpful in expediting the application, he is encouraged to telephone the undersigned at (972) 731-2288.

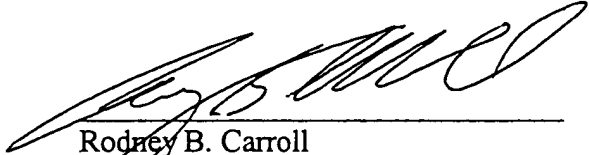
The Commissioner is hereby authorized to charge payment of any further fees associated with any of the foregoing papers submitted herewith, or to credit any overpayment thereof, to Deposit Account No. 50-1515 , Conley Rose, P.C.

Respectfully submitted,

CONLEY ROSE, P.C.

Date: \_\_\_\_\_

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